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SHUFFLER BIAS CORRECTIONS USING CALCULATED COUNT RATES

Jon R. Hurd, Phillip M. Rinard, and Faye Hsue Los Alamos National Laboratory Los Alamos, NM USA 87545

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ABSTRACT

Los Alamos National Laboratory has two identical shufflers that have been calibrated with a dozen U_3O_8 certified standards from 10 g ^{235}U to 3600 g ^{235}U . The shufflers are used to assay a wide variety of material types for their ^{235}U contents. When the items differ greatly in chemical composition or shape from the U_3O_8 standards a bias is introduced because the calibration is not appropriate. Recently a new tool has been created to calculate shuffler count rates accurately, and this has been applied to generate bias correction factors. The tool has also been used to verify the masses and count rates of some uncertified U_3O_8 standards up to 8.0 kg of ^{235}U which were used to provisionally extend the calibration beyond the 3.6 kg of ^{235}U mass when a special need arose.

Metallic uranium has significantly different neutronic properties from the U_3O_8 standards and measured count rates from metals are biased low when the U_3O_8 calibration is applied. The application of the calculational tool to generate bias corrections for assorted metals will be described. The accuracy of the calculational tool was verified using highly enriched metal disk standards that could be stacked to form cylinders or put into spread arrays.

INTRODUCTION

Two identical shufflers at Los Alamos National Laboratory are routinely used to assay the 235 U masses of a wide variety of inventory items. Yet the only certified calibration standards available are a set of U_3O_8 materials with 92.41% enrichments within identical cans; the 235 U masses range from 10 to 3600 g. While these are excellent standards, there are many inventory items with masses greater than 3600 g 235 U and the standards cannot be expected to accurately represent metals, carbides, aluminum alloys, or even oxides with different moisture contents or container sizes.

With only U_3O_8 standards to use, bias in inventory measurements will be very common. Moisture and carbon raise assay results through energy moderation. Metals have reduced assay masses because of increased self-shielding. Aluminum alloys will have assay results biased high because the uranium has reduced self-shielding and the aluminum provides some moderation.

The fabrication of standards more suitable for even a small fraction of all the inventory materials is unlikely because of the large number of materials and the fabrication expense. The alternative is to apply an accurate calculational bias correction technique. Such a technique has been developed and its use is described in this report.^{1,2}

BIAS CORRECTION

The calculations that are used for bias correction are described in detail in Ref. 1. The shuffler's operation is mathematically simulated by the calculations. The Monte Carlo code MCNP is used to calculate the probability that a neutron from the shuffler's ²⁵²Cf source will induce a fission in the item of interest; neutron multiplication within the item is included. For a source that moves during an irradiation, as done in the Los Alamos shufflers, calculations are done at several stationary ²⁵²Cf

positions. The fission probability at any other position is found by interpolation. A correlation is made between the ²⁵²Cf position and the time into an irradiation, giving the fission probability as a function of time. A differential equation is then solved numerically to find the number of delayed neutron precursor nuclei present at the end of an irradiation. The detection efficiency is found with MCNP for delayed neutrons starting within the item. The neutron multiplication caused by the delayed neutrons is also calculated. Finally, the count rate is found from an expression that describes the shuffler's counts over any number of irradiation and counting cycles. The count rates from this process have been shown to be very accurate for well-characterized oxides and metals that are available as test cases.¹

To calculate a bias correction, count rates from the item of interest and from the U₃O₈ standard material of the same mass are needed.

The calibration expression is

$$\mathbf{r} = \mathbf{f}(\mathbf{m}; \{\mathbf{a}\}), \tag{1}$$

where

r = measured count rate,

f() = calibration function,

 $m = deduced^{235}U$ mass, and

 $\{a\}$ = the set of calibration parameters deduced from the calibration standards.

But another type of material with the same 235 U mass will give a different count rate, r', and therefore a biased mass m' will be inferred from Eq. (1).

There are at least three equivalent ways to perform a bias correction. The first is to calculate new unbiased parameters {b}appropriate for the new material. This is essentially calculating a new calibration curve.

$$r' = f(m; \{b\})$$
 (2)

The same mass of material, in this new form, gives a new count rate r' but the new parameters $\{b\}$ lead to the correct mass nevertheless.

Alternatively, a bias correction can be applied to the incorrect mass m' that results from using Eq. (1) on a material differing from the calibration material.

$$r' = f(m'; \{a\})$$
 (3)

The bias correction factor c to m' is of course m/m' but must be found without measuring m. A set of r and m values are known from measurements on standards (with which calculations agree, it is assumed). Another set of r' and m' values can be calculated for the new material. The bias correction factor as a function of the incorrect mass c(m') can be calculated from the m and m' found when r = r'. When the biased situation of Eq. (3) arises, the incorrect mass m' is multiplied by c(m') to give the correct mass m.

The third technique corrects the bias in the count rate before using Eq. (1) unchanged. For a given 235 U mass m in the new material, the incorrect count rate is r', which can be calculated; the count rate for the standard material r is measured or calculated. The bias correction factor d is r/r'. This can be done for many masses to form a bias correction factor as a function of the incorrect rate d(r'). The correct mass is then deduced from Eq. (1) by using the bias corrected rate r = d(r') r'.

CALIBRATION EXTENSION

Calibration standards cover a certain mass range, while the masses of some inventory items may be outside the range. The calculational method can be used to extend the measurements into new mass ranges.

Exactly this situation arose with our U_3O_8 materials. The largest mass in the standards was 3600 g ^{235}U but some similar inventory items had about 5000 g ^{235}U . We also have two U_3O_8 items with declared ^{235}U masses of 6937 and 8022 g, but they are not certified standards and their pedigree is poorly known. It was decided not to rely on these uncertified materials or the newly devised calculational technique alone, but if the two agreed then a provisional extension of the calibration beyond 3600 g ^{235}U would be granted.

The result is summarized in Fig. 1 where the count rates from the calibration standards are shown up to $3600 \, \mathrm{g}^{235} \mathrm{U}$, the count rates from the two uncertified materials are shown at the high-mass end, and the calculated count rates are shown for all masses. The calculations closely match the measured rates of the standards. The matches with the uncertified materials are very good, but not as perfect as those for the standards. Either the declared masses are not quite correct or the limitations of the calculations are magnified at such large masses. In any case, it is also comforting that the calibration curve and the proposed extension is linear from $1000 \, \mathrm{g}$ to $8000 \, \mathrm{g}$. All these factors make it highly likely that the calibration is accurate up to $8000 \, \mathrm{g}^{235} \mathrm{U}$ and this assessment was accepted by the regulators.

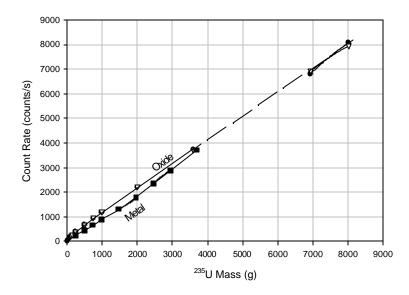


Fig. 1. Measured and calculated count rates are shown for U_3O_8 and HEU metal. The oxide data at and below 3600 g²³⁵U are for certified standards. The dashed portion of the oxide line is the extension of the calibration beyond the certified standards. The black circles are measured rates and the open triangles are calculated rates. The measured and calculated rates for metal are indistinguishable and both are represented by the black squares.

As an aside, it is not possible to extend the calibration to higher masses by stacking cans of U_3O_8 on top of each other or by placing them side by side. For a given ^{235}U mass m in a single can the oxide forms a solid cylinder. The same mass divided into two or more cans has the small masses separated by air gaps and the self-shielding is reduced; the count rate will be well above the true calibration curve. We explored this with pairs of the certified standards and found that the resulting count rates were larger than expected from the correctly extended calibration curve.

An extension was also generated for HEU metal items in a manner very similar to the U_3O_8 case. We had standards in the form of metal disks that could be stacked to create cylinders with masses up to 3700 g 235 U without forming air gaps between the disks. We again had inventory items to measure with even higher masses. The measured calibration curve in this case was not linear, as seen in Fig. 1, so it was even more important that calculations agree with the measurements and then give the shape for higher masses. The calibration curve for metals is below that of oxides because of self-shielding, but the metal curve is poised to cross and be above the oxide curve because of multiplication.

This application was also successful even though the new HEU metallic materials had irregular rather than cylindrical shapes. ²³⁵U masses deduced from the extended calibration curve were within 0.5% to 4.8% of the declared masses. The bias introduced by the shape differences was known to be small from additional calculations that showed only small differences in count rates from widely different shapes as long as the materials were not widely dispersed with air gaps among them.¹

Another example of a bias correction arose when some containers with U_3O_8 mixed with graphite needed to be verified. The graphite is a good neutron moderator and introduces a strong bias in the measurements that use the pure U_3O_8 calibration. Although the density of the material and the existence of other impurities were not known, the calculated count rates agreed with the measured rates to within about 10%, and this was close enough to verify the declared ^{235}U masses.

CONCLUSIONS

We are only beginning to reap the benefits of the new ability to calculate accurate shuffler count rates. We have already been able to measure oxide and metal items with assurance of good accuracy even though physical calibration standards did not exist for the materials in the usual sense. Bias corrections and calibration extensions for shufflers have been needed for many years and the continued application of the new techniques will extend the usefulness of the shufflers.

REFERENCES

- 1. P. M. Rinard, "Calculating Accurate Shuffler Count Rates and Applications," these proceedings.
- 2. P. M. Rinard, "Calculating Shuffler Count Rates," Los Alamos National Laboratory report LA-13815-MS, to be published the summer of 2001.
- 3. J. R. Hurd, R. Siebelist, J. Fitzpatrick, and P. M. Rinard, "Comparison of AWCC and Shuffler Measurements of Uranium Metals," these proceedings.